ADSORBATE INDUCED CONDUCTIVITY OF HYDROGEN-TERMINATED DIAMOND

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Abstract

The origin of the p-type surface conductivity [1, 2] of hydrogen-terminated diamond is not fully understood; however, the electrochemical transfer-doping model proposed by Maier *et al.* [3] is gaining acceptance. According to this model, a p-type accumulation layer forms in the diamond due to the relative positions of the Fermi levels of the diamond and an adsorbed water layer on the surface. The chemical potential of electrons in the water layer is fixed by an electrochemical redox couple, most likely involving dissolved oxygen in equilibrium with air.[4]

$$O_2 + 4H^+ + 4e^- = 2H_2O$$

In our previous work we have studied this effect under different chemical environments and surface terminations.[5] The conductivity increased upon exposure to acidic vapors, which lower the electron chemical potential in the water film, and decreased when exposed to basic vapors, which increase the electron chemical potential. The effect disappeared with fluorine-terminated diamond and oxidized diamond. Organic liquids that wet the hydrophobic diamond surface displace the water film and quench the surface conductivity. Furthermore, transient measurements of conductivity upon imposition of a step temperature change show an immediate increase in conductivity, followed by a decay of conductivity with a much larger time constant. All of these effects are consistent with the electrochemical transfer-doping model.

In order to obtain further insight into the electrochemical transfer-doping model the contact angle and zeta potential were measured at different pH.[6] As the pH is lowered, the model predicts an increase in the concentration of holes in the diamond and the counter-anions in the adsorbed film. At lower pH one would therefore anticipate increased electrostatic attraction between the hole accumulation layer and the negative counter ions in the water film and therefore a decreased contact angle. The increased electron transfer from the diamond to the water film at low pH should also reveal itself in a more positive zeta potential (more positive surface charge) at low pH. Furthermore, these effects should be more pronounced for hydrogen-terminated compared to oxygen-terminated diamond.

Contact angles of hydrogenated samples decreased uniformly as the pH was lowered from pH = 7 to pH = 1. Reduction of the concentration of electrochemical acceptors, *i.e.*, the dissolved oxygen, should decrease the electron transfer from the diamond and reduce the effect of pH on contact angle. Reduction of dissolved oxygen was accomplished by addition of sodium bisulfite. The effect of pH on contact angle essentially disappeared at both low and high pH after the addition of excess sodium bisulfite. Oxidized diamonds showed a decrease in contact angle as the pH was increased above 3. This latter effect is attributed to ionization of oxygen

containing functional groups on the surface. Zeta potentials of both oxygen and hydrogen-terminated diamond were measured at different pH. At high pH, both surface terminations show a negative surface charge (See Fig.1). However, the isoelectric point for hydrogen-terminated diamond occurred at approximately pH = 5.2, below which pH the surface charge is positive. In contrast, oxidized diamond did not show a positive zeta potential until the pH was reduced to 1.2.

Although the results, in general, support the electrochemical transfer-doping model, significant uncertainty remains. In particular, a greatly enhanced effect is seen with rectifying contacts compared to ohmic contacts.[5] Understanding of this effect will require detailed modeling of the diamond/contact/film interface.

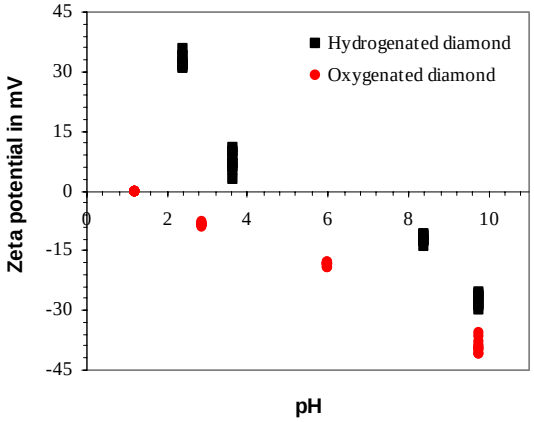

Figure 1. Zeta potential as a function of pH for both hydrogen and oxygen terminated diamond.

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